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ELECTRONICS LABORATORY, Syracuse, New York

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SEMIANNUAL REPORT

For the Period January 1, 1964 - June 30, 1964

AROMATIC ORGANIC LASER DEVELOPMENT

by

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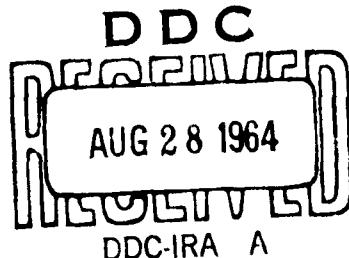
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Prepared for

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Department of the Navy
Washington 25, D.C.



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GENERAL  ELECTRIC

Semiannual Report

AROMATIC ORGANIC LASER DEVELOPMENT

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from the
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I. INTRODUCTION AND SUMMARY

The purpose of this program has been to produce coherent stimulated emission in the blue-green region in a purely organic dopant-host system. Towards this end, the experimental program of the last six months has been concerned primarily with the study of the optical properties of the polymers to be employed and on the development of a high peak power pumping source. Progress has been made in both of these areas.

II. MATERIALS SELECTION AND PREPARATION

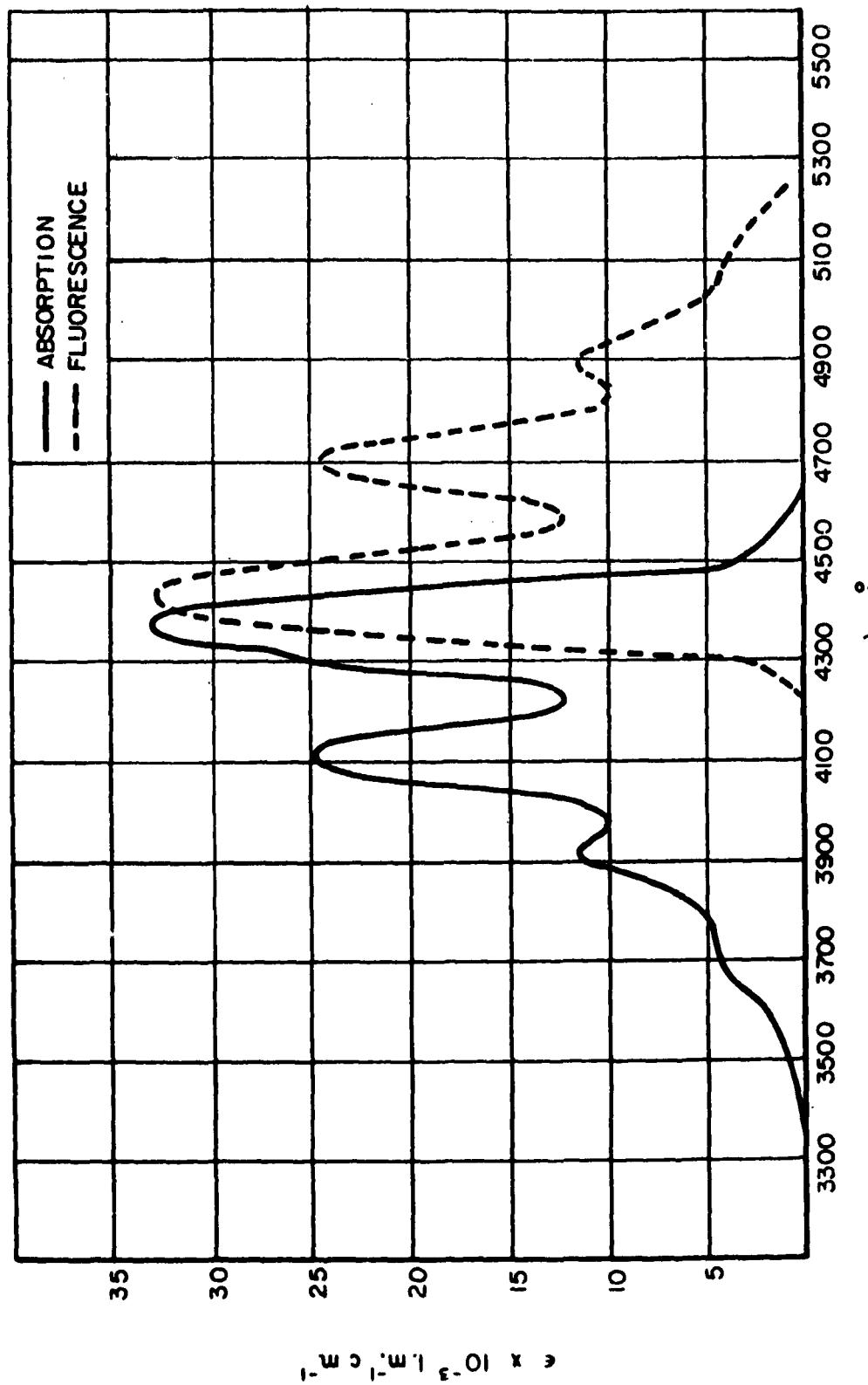
A. Spectroscopic Measurements

The previous semiannual report noted that two molecules had been found with suitable spectroscopic properties. The two compounds are perylene and 9-aminoacridine. The routine spectroscopic features of these compounds were presented in the previous report. We have made some additional spectroscopic measurements. From the absorption and emission data for perylene, shown in Figure 1, the terminal-ground state splitting is measured by the energy difference between the 4710 Å fluorescence peak and the crossover (the 0-0 transition) point between the absorption and emission spectrum at 4445 Å. The energy separation is 1266 cm^{-1} . The long wavelength absorption ($> 4445 \text{ Å}$) of the perylene is due to transitions of the upper vibrational-rotational levels of the ground electronic state to the lowest vibrational-rotational level of the excited state. The population arises from simple Boltzmann statistics. Owing to the near perfect mirror symmetry of the absorption and fluorescence spectrum, it would be anticipated that absorption at the 4710 Å terminal state would have the same cross section as the 4100 Å peak.

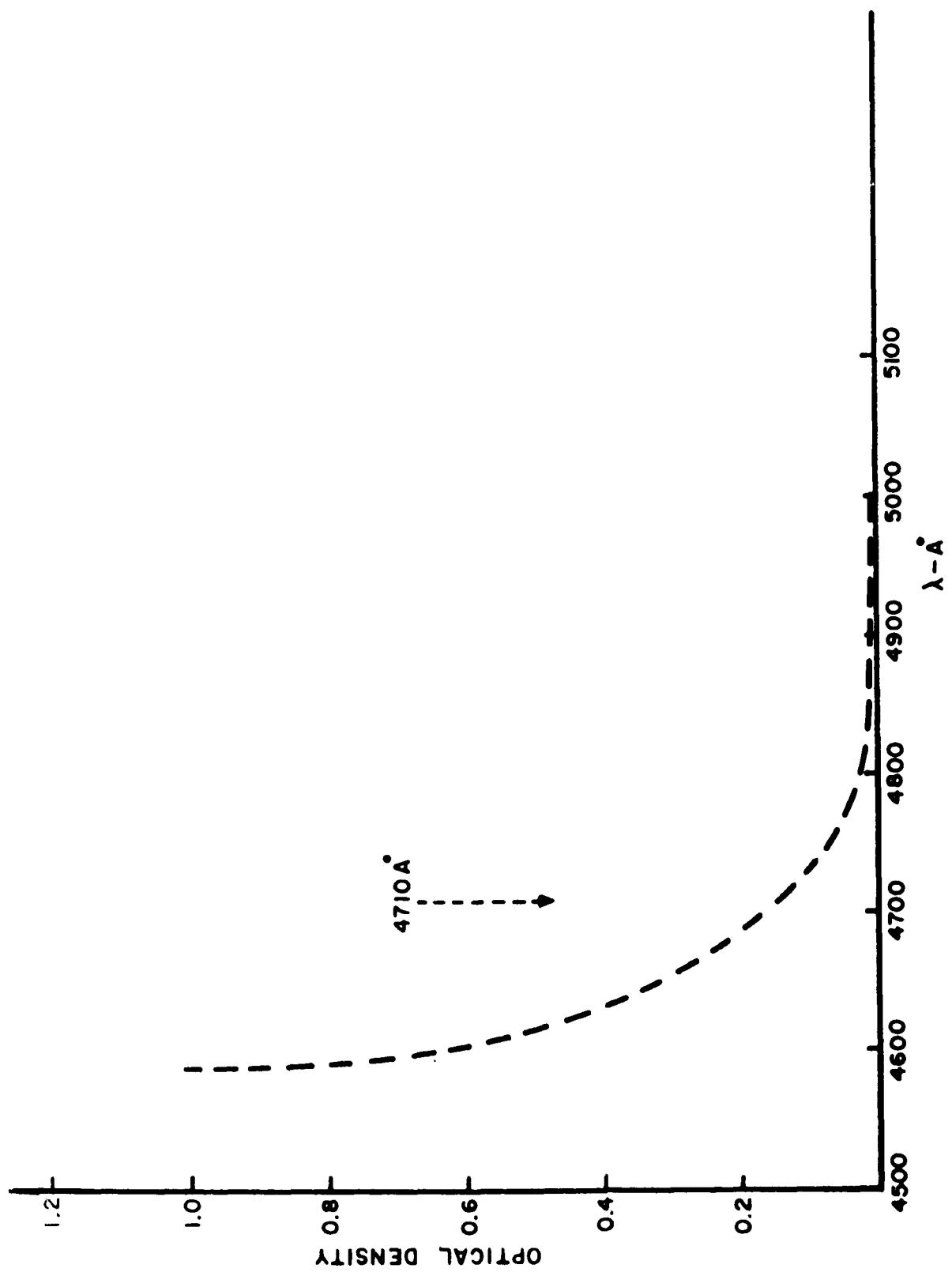
Figure 2 illustrates the experimentally obtained absorption of a 5.56×10^{-4} molar solution of perylene in benzene using a 10 cm cell. From the optical density at 4710 Å, we calculate that the terminal ground state separation is about 1300 cm^{-1} in fair agreement with the spectroscopic value. Due to the assumptions employed, no better agreement should be expected.

B. Polymerization Studies

In the previous report, it was shown that casting of the dye-monomer solution in the absence of oxygen produced the highest optical quality polymer rods. When the birefringence studies were begun, it was found necessary to add dibutyl phthalate (see Section C). the polymerization rates in the presence of the dibutyl phthalate were slowed down to the



ABSORPTION AND FLUORESCENCE SPECTRUM OF PERYLENE IN BENZENE
FIGURE 1



LONG WAVELENGTH ABSORPTION OF 5.56×10^{-4} MOLAR PERYLENE IN BENZENE (10cm PATH LENGTH)

FIGURE 2

extent that the organic dyes were oxidized by the catalyst, benzoyl peroxide. It was not found possible to adjust the rates of growth and concentration of catalyst used to prevent degradation of the dye. Part of the difficulty can be traced to the fact that the catalyzing agent is not completely used up at the end of the polymerization.

Another catalyst has been found which is used up completely, requires lower initial concentrations and does not oxidize the dye. This catalyst is α, α' - azodiisobutyronitrile. The appropriate samples have now been prepared for the stress optical measurements to be described below. The samples are grown in oxygen evacuated pyrex tubes under the temperature cycle given below;

40°C	2 days
Raise 40° to 60°C	1 day
Raise 60°C to 100°C	1 day
100°C	3 days
Cool and anneal at 102°C.	

C. Optical Properties of Polymers

It was noted in the last semiannual report that most polymers are birefringent in their natural state due to stresses which cannot be annealed out of the samples. If a high polymer is stressed there is a restoring force which originates from two types of molecular processes. One of these is an entropy component which is the result of the orientation produced by the deformation. Brownian motion of each segment of the distorted polymer chain operates to return the chain to its more probable configuration. The entropy force increases with temperature because of increased Brownian motion. There is also a potential energy component of the force which is a result of the attraction (or repulsion) between molecules that have been pulled apart (or squeezed together) during the deformation. Thermodynamically, the force, f , is

$$f = \left(\frac{\partial W}{\partial l} \right)_T = \left(\frac{\partial U}{\partial l} \right)_T - T \left(\frac{\partial S}{\partial l} \right)_T$$

For an ideal rubber, only the entropy contribution is present.

If a polymer is stretched and held at constant length, the stress relaxes as a result of a rearrangement of the chains from their initial deformed configuration to the more probable configuration. Chemical reactions, viscous flow, localized strain rearrangement and crystallization are some of the processes which can change the polymers' configuration.

High polymers typically exhibit two types of behavior. At, or near, room temperature, most of them are in a glassy state characterized by frozen chain orientations. For polymers in this state, the birefringence is proportional to the stress for small values of stress and the stress arises principally from the potential energy term described above. At higher temperatures, the polymer changes conformation to a rubbery state characterized by stresses arising almost from the purely entropy considerations. A more complete discussion of these effects can be found elsewhere⁽¹⁾.

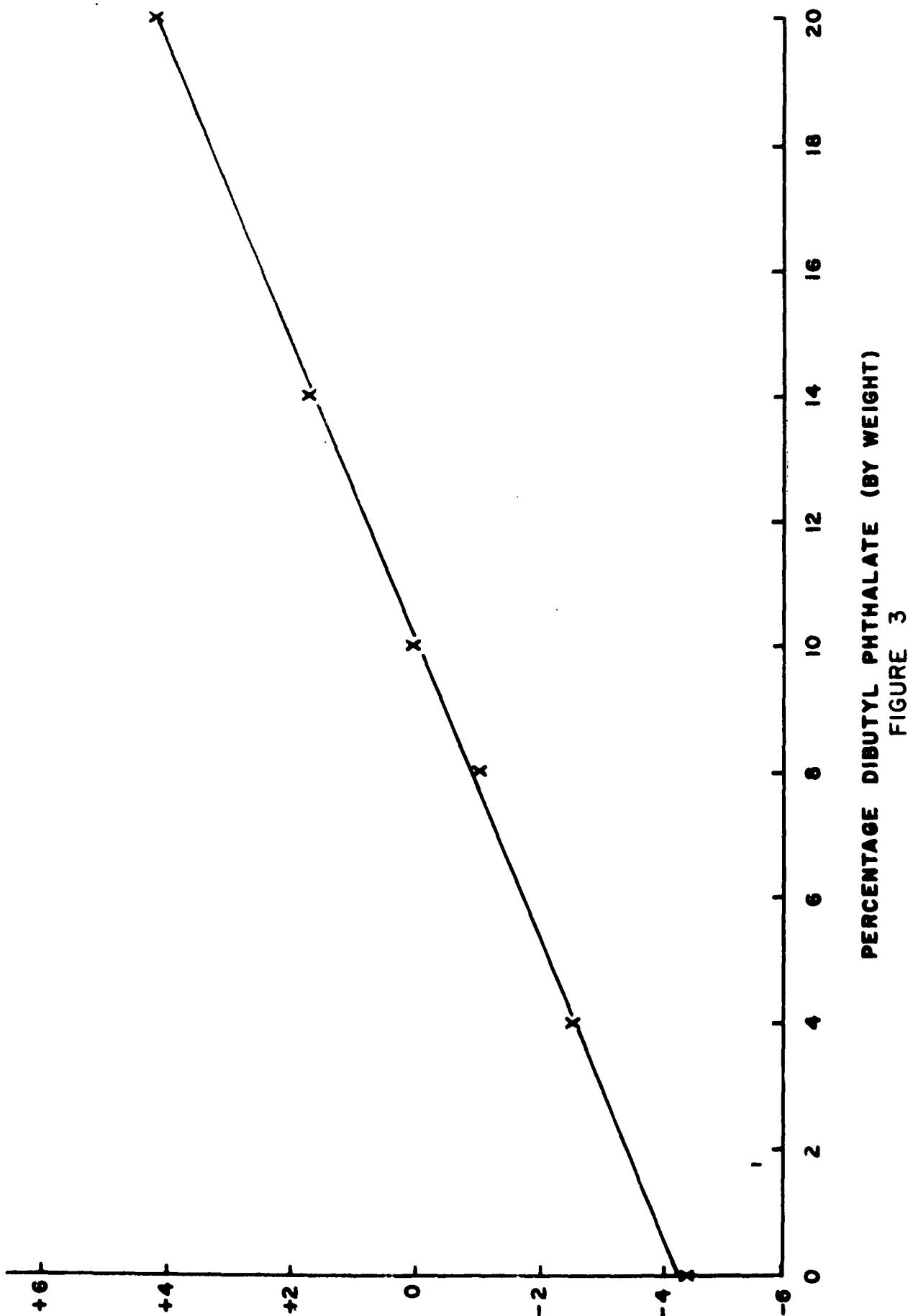
The transition from the rubbery state to the glassy state is a second-order transition. Since all polymers of interest must be annealed in the rubbery state and cooled to a glassy state, and since it is not possible to maintain exact thermal equilibrium in passing through the transition temperature, it is not possible to remove all the stresses in a polymer sample. As a result, all polymers exhibit stress-induced birefringence, even after careful annealing.

The birefringence is related to the stress by

$$\Delta N = KF$$

where K is the stress-optical coefficient and
 F is the force per unit area.

Typical values of K for polymethyl methacrylate are $-4 \times 10^{-13} \text{ cm}^2/\text{dyne}$. It has been shown by Lamble and Dahmouch⁽²⁾ that it is possible to obtain a zero stress-optical coefficient in polymethyl methacrylate by the addition of dibutyl phthalate. Figure 3 shows a measured stress-optical coefficient



for the modified polymer as a function of composition measured at 5461 Å. This was made on the benzoyl peroxide-catalyzed form of the undoped polymer. Due to the necessary changes in the polymerization of the polymer described in Section B, it will be necessary to re-determine the stress-optical coefficients for the polymers produced using the new catalyst system. The measurements will be made at 5461 Å and 4358 Å in order to ensure that differences in dispersion between the ordinary and extraordinary ray do not require a slight shift in composition. When this is done, it will be possible to produce a polymer which is optically isotropic and thus suitable for forming a proper resonant cavity.

III. DEVICE DEVELOPMENT

The development of a suitable optical pump involves generally a pulse of at most 1 microsecond duration, with a peak power of at least 100 megawatts without any sacrifice in terms of brightness of a lamp. It has become increasingly evident that black body systems such as the common xenon flash tube exhibit saturation of the lamp brightness at black body temperatures of 12-15,000°K⁽³⁾. Previous experiments with the generation of short pulses of 100-200 nanoseconds duration showed that the lamps did not increase in brightness appreciably with apparent increased peak power. The theta-pinch lamp work described by Feldman and coworkers⁽⁴⁻⁶⁾ is the subject of a current controversy⁽⁷⁾. However, it is a lamp which yields a pulse of short duration and whose brightness remains relatively high. During this report period, we have constructed such a lamp and have attempted to maximize the various operating parameters.

The lamp-capacitor system utilizes a 0.9 microfarad, 50 KV clam-shell capacitor (GE 14F756G2) in conjunction with a single turn coil surrounding a toroidal bottle containing argon at various pressures with an appropriate switch. The bottle is 1 1/8" OD, 5/16" ID, and 3" long. The fill gas pressures range from 1-100 millimeters.

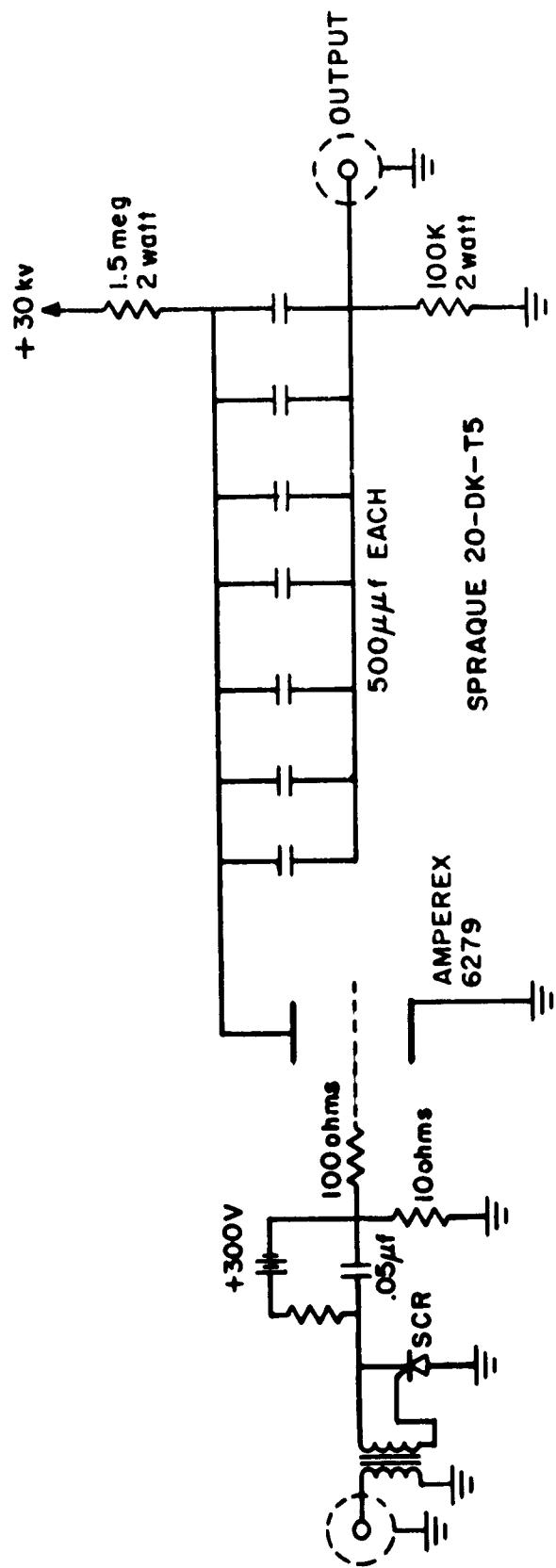
The chief experimental difficulty involves the switch. A number of switches have been tried. Initially, a high current ignitron was tried but the inductance (20 to 30 nanohenries) limited the current rise, so that inefficient operation and a rather long pulse of about 10 microseconds was attained. A simple airgap switch proved somewhat more satisfactory but at the higher voltages required, the gap separation was too large to switch the gap rapidly. Pulses of 4-5 microseconds were obtained. A switch of the Lupton design⁽⁸⁾ first made at NRL was constructed and it gave further improved performance. The switch now being tested is a pressurized version of the Lupton design manufactured by Cooke Engineering Company of Alexandria, Virginia (Model ME40-10,000).

It is capable of fast triggering at 50 KV without a wide gap separation. The characteristics of this gap are as follows:

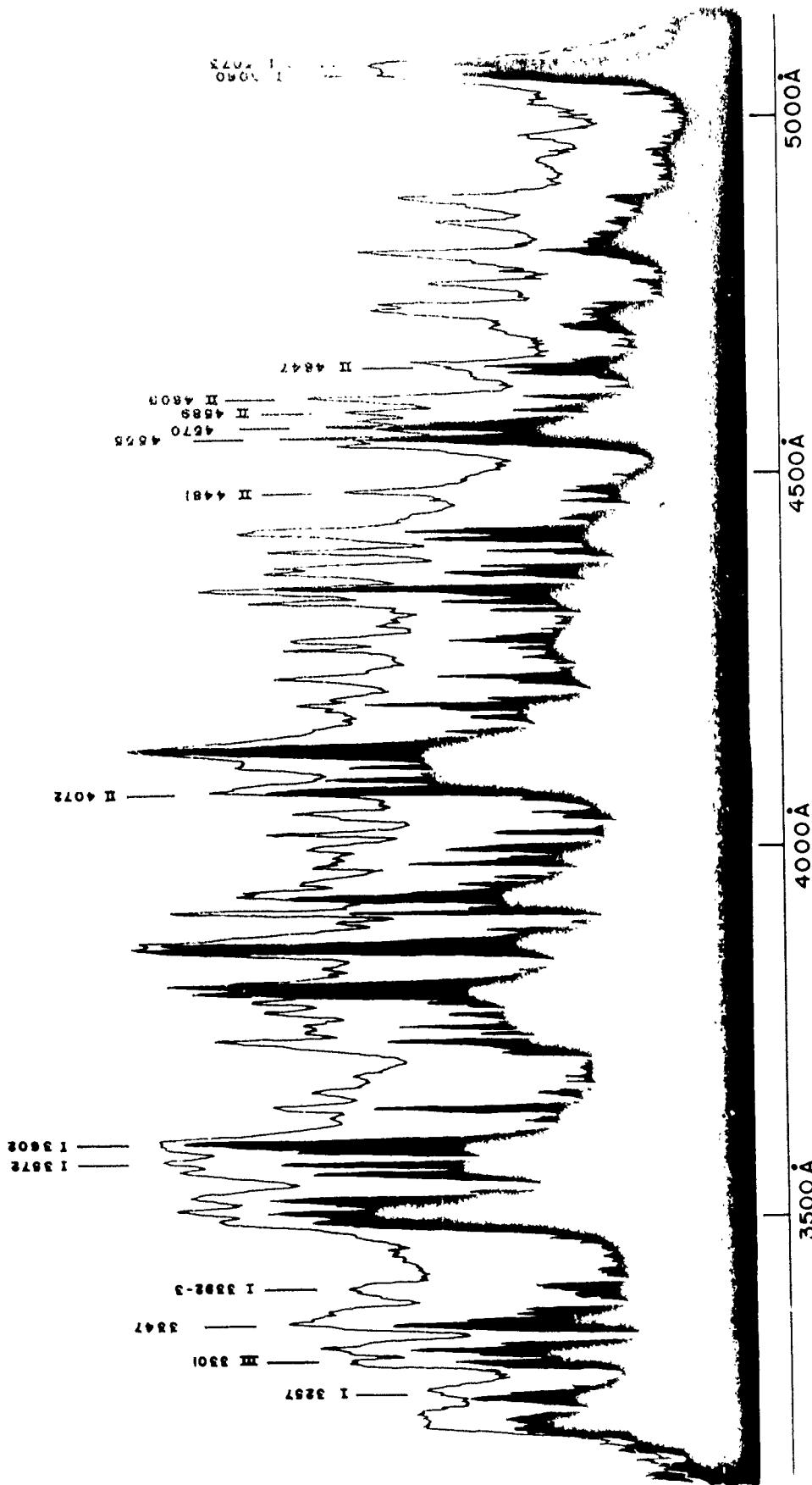
maximum voltage	-	50 KV
peak current	-	10^6 amperes
total energy switched	-	10^4 joules
inductance	-	less than 10 nanohenries
dynamic impedance	-	5 milliohms

In the circuit we employ this should produce a pulse which rises in about 1.5×10^{-7} seconds and lasts about 1 microsecond depending on the coupling efficiency. The importance of rapid switching cannot be over-emphasized. In order to trigger the switch properly, a voltage pulse rising at the rate of 1 KV/nanosecond is required. Figure 4 shows the appropriate thyratron circuit which is wired in accordance with good high frequency practice.

The spectral characteristics of this lamp are important. Feldman's data⁽⁷⁾ is shown in Figure 5. The black spectrum is for an argon gas pressure of one millimeter and the white spectrum is at 20 millimeters gas pressure. At 20 millimeters, the continuum is obviously more significant. We plan to perform similar experiments at General Electric's Advanced Technology Laboratories using equipment developed under a previous flash lamp study contract (NONR-4121(00)). These experiments will be performed in order to measure the absolute brightness and efficiency of the source in addition to its time resolved spectroscopic characteristics.



SPARK GAP FIRING CIRCUIT
FIGURE 4



RING DISCHARGE SPECTRA IN ARGON

FIGURE 5

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